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Report on Task 2.3: Physical Properties Measurement To Lawrence Livermore National Laboratory for Contract B345772

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Task 2: Performance Testing

Report on Task 2.3: Physical Properties Measurement

1 Summary

This report contains a summary of the results generated for Task 2.3: Physical Properties Measurement. The aim of this task was to determine the theoretical density of selected samples and from this determine an approximate relationship between open and closed porosity.

2 Experimental

2.1 Samples Tested

Three Pu-doped samples (Table 1) were chosen from Task 1.2 work [1] for use in determining the theoretical density:

1. Oxide-route wet milled baseline composition (B1-1) sample (Pu68). This was chosen because it represents the baseline composition. An SEM micrograph of the sample is given in Appendix A figure A-1 and the EDS results for this sample are given in Table A-1.

2. Oxide-route wet milled baseline composition + impurities (A-7) sample (Pu75). This was chosen because it represents the baseline composition with typical impurities added. An SEM micrograph of the sample is given in Appendix A, figure A-2 and the EDS results for this sample are given in Appendix A, Table A-2.

3. Alkoxide-route wet milled "nominally" 10 % perovskite composition (B1-13) sample (Pu105). This was chosen because it was composed almost entirely of pyrochlore, with only a small amount (1 – 2 vol. %) of rutile present. An SEM micrograph of the sample is given in Appendix A, figure A-3 and the EDS results for this sample are given in Appendix A, Table A-3.

2.2 Determination of Theoretical Density of the Three Pu-doped Samples

The lattice parameters were determined from x-ray diffraction patterns. From these the unit cell volumes of the phases were calculated as follows:

- For pyrochlore (cubic), $V = a^3$
- For brannerite (monoclinic), $V = abc \sin \beta$
- For 2M zirconolite (monoclinic), $V = abc \sin \beta$
- For rutile (tetragonal), $V = a^2c$

where: V = the unit cell volume; *a*, *b*, and *c* are the lattice parameters; and, β is the unit cell angle.

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[1] M W A Stewart, E R Vance, R A Day and A Brownscombe, *Interim Report on Task 1.2: Near Equilibrium Processing Requirements*, ANSTO Report No. R99m012, 1 April 1999, Materials Division, ANSTO, Lucas Heights, Australia.

The EDS results from Task 1.2 (Tables 2-4) were used to determine the formula weight of each phase and hence the mass contained within a unit cell. The theoretical density of each phase was calculated from these two measurements.

Due to the paucity of XRD peaks recognisable as being due to Hf-doped rutile (Table 5) only the *a* lattice parameter could be determined. Therefore the *c* lattice parameter was determined by assuming that it increased by the same amount, relative to undoped rutile, as the *a* lattice parameter.

For the phases that were present in amounts too small to measure, the lattice parameters used were taken from published data and the densities were estimated using the EDS analyses.

Image analysis of the SEM micrographs (Appendix A), utilising a computerised pixel counting technique, was used to determine the volumetric amount of each phase present in the sample.

Knowing the volumetric amount of each phase present, and their densities, a theoretical density was calculated for each composition.

2.3 Estimation of the Theoretical Density of the Th/U-doped Samples

The lattice parameters calculated for the Pu-doped samples were assumed to remain unchanged for the Th-doped samples[2]. The theoretical densities of the Th-doped samples were estimated from the measured densities of the Pu-doped samples by substituting Th for Pu in the composition formulation and assuming that the phase composition remained constant.

2.4 Determination of the Relationship Between Open and Closed Porosity

The values of the theoretical density were used to determine the open and closed porosity of the samples. From this plots of porosity versus density can be made and the relationships estimated by curve fitting methods.

3 Results and Discussion

3.1 Calculated Lattice Parameters

The calculated lattice parameters are given in Table 2. From these the cell volumes were calculated, as given in section 2.2.

3.2 Phase Analysis of Pu-doped Samples

Compositional analysis, as determined by image analysis of the Pu-doped samples was determined by phase analysis and is given in Table 3.

[2] We intend to determine the lattice parameters of selected Th-doped samples later this year.

Table 2: Calculated Lattice parameters for the Pu-doped samples.

Phase	Pu68 (B1-1)	Pu75 (A-7)	Pu105 (B1-13)	Published data for end members
	Baseline	Baseline + Impurities	"Nominally" 10 % perovskite (Pyrochlore + Rutile*)	
	Lattice Parameters (angstrom)			
Pyrochlore	$a = 10.1461$	$a = 10.1429$	$a = 10.1329$	$a = 10.1625$ +
Brannerite	$a = 9.8223$ $b = 3.7292$ $c = 6.8645$ $\beta = 118.710°$	$a = 9.8340$ $b = 3.7274$ $c = 6.8594$ $\beta = 118.614°$		$a = 9.8016$ $b = 3.7620$ $c = 6.9125$ $\beta = 118.97°$ @
2M Zirconolite		$a = 12.524$ $b = 7.231$ $c = 11.365$ $\beta = 100.583°$		
Rutile **	$a = 4.615$ $c = 2.9732$	$a = 4.610$ $c = 2.9700$	***	$a = 4.5933$ $c = 2.9592$ #

* "Nominally" 10% perovskite batch.

** *c* is estimated based on same percentage increase in the *a* lattice parameter of Hf-doped rutile relative to undoped rutile[3].

*** Not measured; assumed to be the same as Pu68 for density calculations.

\+ $CaUTi_2O_7$ JCPDS card 42-0425.

@ UTi_2O_6 JCPDS card 12-0477.

\# TiO_2 JCPDS card 21-1276.

[3] A sample of Hf-doped rutile is currently being prepared to test the validity of this assumption.

Table 3: Composition analysis results for the Pu-doped samples.

Phase	Pu68 (B1-1)		Pu75 (A-7)		Pu105 (B1-13)	
	Baseline		Baseline + Impurities		"Nominally" 10 % Perovskite (Pyrochlore + Rutile)	
	Composition (Vol. %)					
	With Pores	Solid	With Pores	Solid	With Pores	Solid
Pyrochlore	73.7	79.3	62.8	66.3	88.8	98.3
Brannerite	8.7	9.4	12.3	13.0		
2M Zirconolite			13.4	14.1		
Rutile	10.4	11.2	5.7	6.0	1.5	1.7
$(Pu,U)O_2$	0.1	0.1	0.1	0.2		
Silicate			0.4	0.4		
Porosity	7.1	-	5.3	-	9.7	
Total	100.0	100.0	100.0	100.0	100.0	100.0.0

3.3 Calculated Densities of Phases in the Pu-doped Samples and Calculated Theoretical Densities of Selected Pu-doped Samples

The densities of the individual phases (calculated and estimated) and the calculated theoretical densities for the three Pu-doped samples are given in Table 4. Some variation in the density of each phase occurs across the compositions, e.g., the density of the pyrochlore in Pu105 is lower because it has more Ca present, at the expense of the heavier Gd.

3.4 Actual Measured Densities of the Pu-doped Samples

The densities of samples made from the same composition are given in Table 5.

There are some variations between the porosities measured from the SEM micrographs and this is probably indicative of the sampling error from the micrograph.

- Pu68 (sintered 1350°C/4h/Ar)- the measured bulk density was 5.575 g/cm^3 (94.7 % theoretical density), with an apparent porosity of 0.1 %, a closed porosity of 5.2 %, and a total porosity of 5.3 %. From the micrograph the measured porosity was 7.1 %.

- Pu75 (sintered 1325°C/4h/Ar)- the measured density was 5.564 g/cm^3 (93.8 % theoretical density), with an apparent porosity of 0.0 %, a closed porosity of 6.2 %, and a total porosity of 6.2 %. From the micrograph the measured porosity was 5.3 %.

- Pu105 (sintered 1350°C/4h/Ar)- the measured density was 4.98 g/cm^3 (85.2 % theoretical density), with an apparent porosity of 1.0 %, a closed porosity of 13.8 %, and a total porosity of 14.8 %. From the micrograph the measured porosity was 9.7 %.

Table 4: Calculated densities of each phase and the theoretical density of the selected Pu-doped samples.

Phase	Pu68 (B1-1)	Pu75 (A-7)	Pu105 (B1-13)	Published Phases
	Baseline	Baseline + Impurities	Pyrochlore + Rutile	
	Density (g/cm^3)			
Pyrochlore	5.994	6.019	5.869	6.145 #
Brannerite	6.218	6.176		6.401 $
2M Zirconolite		5.856		
Rutile	4.856	4.705	*4.712*	4.250 +
$(Pu,U)O_2$	*10.86* *	*10.75* *		11.668 (PuO_2) &
Silicate		*3.0*		
Calculated Theoretical Density	5.890	5.931	5.850	

* Figures in italics are estimates used to calculate the theoretical density. They have been calculated using the phase composition as measured by EDS.

\# $CaUTi_2O_7$, JCPDS file 42-425.

$ UTi_2O_6 (brannerite), JCPDS file 12-477.

\+ TiO_2 (rutile), JCPDS file 21-1276.

& PuO_2, JCPDS file 41-1170.

Table 5: Measured densities of Pu-doped samples

Sample	Sintering Temp (°C)	Sintering Atm.	Bulk Density (g/cm^3)	Theoretical Density (%)	Apparent ("open") Porosity (%)	Closed Porosity (%)	True Porosity (%)
Pu67 Composition B1-1 Oxide-route, Dry Milled							
Pu67	1275	air	5.362	91.0	2.9	6.1	9.0
Pu67	1275	Ar	5.238	88.9	2.6	8.5	11.1
Pu67	1300	Ar	5.374	91.3	0.4	8.4	8.8
Pu67	1325	Ar	5.342	90.7	1.4	7.9	9.3
Pu67	1350	Ar	5.397	91.6	1.3	7.1	8.4
Pu67	1375	air	5.402	91.7	2.2	6.1	8.3
Pu67	1375	Ar	5.450	92.5	1.8	5.7	7.5
Pu67	1375	Ar	5.431	92.2	2.2	5.6	7.8
Pu67 #	1400	air	3.954	67.1	31.0	1.9	32.9
Pu67	1400	air	5.310	90.2	3.7	6.2	9.9
Pu67	1400	Ar	5.415	91.9	1.4	6.7	8.1
Pu67	1400	Ar	5.479	93.0	0.0	7.0	7.0
Pu68 Composition B1-1 Oxide-route, Wet Milled							
Pu68	1275	air	5.556	94.3	0.0	5.7	5.7
Pu68	1275	Ar	5.581	94.8	0.1	5.3	5.3
Pu68	1300	Ar	5.610	95.3	0.0	4.8	4.8
Pu68	1325	Ar	5.603	95.1	0.8	4.9	4.9
Pu68	1350	Ar	5.575	94.7	0.1	5.2	5.3
Pu68	1375	air	5.442	92.4	1.8	5.8	7.65
Pu68	1375	Ar	5.547	94.2	0.8	5.0	5.8
Pu68	1375	Ar	5.567	94.5	0.7	4.8	5.5
Pu68 #	1400	air	3.963	67.3	28.5	4.2	32.7
Pu68	1400	air	5.364	91.1	0.5	8.4	8.9
Pu68	1400	Ar	5.402	91.7	1.0	7.3	8.3
Pu68	1400	Ar	5.531	93.9	0.1	6.0	6.1
Composition A-7 Alkoxide-route, Wet Milled							
Pu71	1300	Ar	4.96	83.6	0.02	16.2	16.4
Composition A-7 Oxide-route, Wet Milled							
Pu75	1325	Ar	5.564				
Pu75	1325	Ar	5.540	93.4	0.0	6.6	6.6
Composition B1-13 Oxide-route, Wet Milled							
Pu101	1350	Ar	5.284	90.3	0.9	8.8	9.7
Composition B1-13 Oxide-route, Dry Milled							
Pu102	1350	Ar	5.484	93.6	0.0	6.4	6.4
Composition B1-13 Alkoxide-route, Wet Milled							
Pu105	1350	Ar	4.983	85.2	1.0	13.8	14.8

Problems occurred during the sintering run and it is suspected that samples did not reach the set sintering temperature.

3.5 An Estimate of the Errors in Measurement

Measurement errors occur in several of the analyses undertaken:

1. EDS results have a typical standard deviation on each phase of ~ 0.35 %, which gives a 95 % confidence interval of ~ 0.7 %.

2. Errors in the lattice parameter measurements are confined to the fourth decimal place.

3. Errors in the compositional analysis are typically 1 - 2 %, though larger systematic errors could occur, due to, for example, the use of unrepresentative SEM micrographs. In an extreme case, e.g., overestimation of the amount of rutile and underestimation of the amount of brannerite by 1 %, these would lead to a decrease in the estimated density by 0.02 g/cm^3 (equivalent to 0.3 % of 5.9 g/cm^3).

Thus the error in the theoretical density determination would be expected to be ~ 1 % or ~ 0.05 g/cm^3.

3.6 Estimated Theoretical Densities of the Th-doped Compositions

Theoretical densities of the Th doped samples have been calculated from the calculated Pu-doped theoretical densities with Th substituted for Pu, using constant lattice parameters (see section 2.3). The results are given in Table 6.

Table 6: Estimated theoretical densities of Th-doped compositions

Composition	Pu-doped composition calculated theoretical density (g/cm^3)	Estimated Th-doped composition theoretical density (g/cm^3)
Baseline	5.89	5.87
Baseline + Impurities	5.93	5.91
Nominally 10 % pyrochlore (Pyrochlore + Rutile)	5.85	5.83

3.7 Measured Densities of the Th-doped Compositions

The measured densities of the Th-doped samples prepared by sintering in Ar or air are given in Table 7.

Table 7: Measured densities of samples made from oxide-route wet and dry milled batches.

Sample No.	Milling Method	Sint. Temp. (°C)	Sint Time (h)	Sint. Atm.	Density (g/cm^3)	Density Theoretical (%)	Open Porosity (%)	Closed Porosity (%)	Total Porosity (%)
Composition	B1-14								
mws980259	Dry	1350	4	Ar	4.50	77.2	22.8	0.0	22.8
mws980260	Dry	1350	4	Ar	4.58	78.6	19.9	1.5	21.4
mws980265	Dry	1350	4	air	3.76	64.5	32.9	2.6	35.5
mws980266	Dry	1350	4	air	3.83	65.8	31.9	2.4	34.3
mws980283	Dry	1350	4	Ar	4.18	71.7	27.3	1.1	28.3
mws980284	Dry	1350	4	Ar	4.00	68.7	30.8	0.5	31.3
mws980287	Dry	1350	4	air	4.06	69.6	25.4	5.0	30.4
mws980288	Dry	1350	4	air	4.22	72.4	24.2	3.4	27.6
mws980257	Wet	1350	4	Ar	5.10	87.6	3.1	9.4	12.4
mws980258	Wet	1350	4	Ar	5.18	88.9	1.5	9.6	11.1
mws980263	Wet	1350	4	air	4.78	82.0	6.4	11.6	18.0
mws980264	Wet	1350	4	air	4.88	83.7	5.2	11.1	16.3
mws980281	Wet	1350	4	Ar	4.85	83.2	6.4	10.4	16.8
mws980282	Wet	1350	4	Ar	4.88	83.7	4.9	11.4	16.3
mws980285	Wet	1350	4	air	5.13	88.0	0.8	11.2	12.0
mws980286	Wet	1350	4	air	5.12	87.8	2.0	10.2	12.2
mws980397	Wet	1350	75	Ar	5.32	91.3	1.0	7.7	8.7
mws980411	Wet	1400	4	Ar	5.28	90.5	2.3	7.2	9.5
Composition	B1-2								
mws980134	Dry	1350	4	Ar	4.70	80.1	17.9	2.0	19.9
mws980139	Dry	1350	4	Ar	4.91	83.6	13.7	2.7	16.4
mws980140	Dry	1350	4	Ar	4.92	83.9	13.8	2.4	16.1
mws980133	Wet	1350	4	Ar	5.04	85.9	11.4	2.7	14.1
mws980137	Wet	1350	4	Ar	5.62	95.8	0.5	3.7	4.2
mws980138	Wet	1350	4	Ar	5.57	94.9	0.2	5.0	5.1
mws980391	Wet	1350	75	Ar	5.41	92.2	5.1	2.2	7.9
mws980407	Wet	1400	4	Ar	5.44	92.7	1.6	5.7	7.3
Composition	B1-4								
mws980151	Dry	1350	4	Ar	4.97	84.1	10.5	5.4	15.9
mws980152	Dry	1350	4	Ar	5.19	87.8	7.9	4.3	12.3
mws980149	Wet	1350	4	Ar	5.19	87.8	0.6	11.6	12.2
mws980150	Wet	1350	4	Ar	5.28	89.4	0.4	10.1	10.6
mws980394	Wet	1350	75	Ar	4.99	84.5	3.7	11.8	15.5
mws980408	Wet	1400	4	Ar	5.11	86.5	1.7	11.8	13.5

3.8 Relationship Between Open and Closed Porosity

3.8.1 Samples made from Pu-doped compositions

The results given in Table 5, showing porosity (total (true), apparent and closed) versus density from the samples made from batches Pu67 (dry milled baseline) and Pu68 (wet milled baseline) have been plotted (figure 1). There is some scatter in the results and this may reflect the limitations

of the measurement techniques used for porosity measurement[4]. There is a lack of data in the middle porosity range. The results follow the trend typical in ceramics with the closed porosity increasing as the density increases above 90 % TD and the apparent porosity getting very small (< 1 %) between 92 and 96 % theoretical density.

An approximate relationship between porosity and density is given in figure 2.

The apparent porosity versus closed porosity has been plotted (figure 3). However the scatter of the results, plus the lack of data over certain apparent porosities, has meant that the relationship, in terms of fitting a model trend curve, cannot be evaluated accurately.

3.8.2 Samples made from Th-doped compositions

The results from the samples for Th-doped compositions given in Table 7 have been plotted as open and closed porosity versus (figure 4). There is some scatter in the results (as discussed above). The trends are similar to those of the Pu-doped samples. Samples made from composition B1-4 reach apparent porosities of < 1 % at ~ 90 % theoretical density

Approximate relationships between the apparent porosity and density is given in figures 5 (combined results) and 6. Linear regressions have been plotted for the individual compositions (figure 6). It can be seen that each composition has a different relationship between apparent porosity and density. B1-2 reaches "closed" porosity at ~ 95 % theoretical density. B1-14 and B1-4 reach closed porosity at ~ 90 % theoretical density. This difference is probably due to changes in composition, which in turn change the sintering behaviour, e.g., B1-4 will have a glassy phase which is liquid at the sintering temperature and consolidates via liquid phase sintering, whereas B1-2 is glass free and sinters by solid state reaction.

3.9 Future Proposed Work

As part of the next stage of work for Task 1.2 it has been proposed that the compositions originally examined via wet milling, dry milling and alkoxide-routes be reassessed using attrition milling. If this work proceeds then it is proposed that the samples from these tests be examined, as above, to determine the relationship between apparent and closed porosity under these processing conditions.

The lattice parameters of selected Th-doped samples will be determined.

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[4] The use of Archimedes' method to measure the porosity of small samples has some limitations. Standards such as ASTM C373-88, *Standard Test Method for Water Absorption, Bulk Density, Apparent Porosity and Apparent Specific Gravity of Fired Whiteware Products,* or Australian Standard AS1774.5-1989, *Refractories and refractory materials - Physical test methods, Method 5: The determination of density, porosity and water adsorption were* originally designed for samples > ~ 50 g measured to 0.01 g. The Th-doped and Pu-doped test samples in this work are typically 0.5 to 1 g. The inaccuracies with small sample size can be overcome by more precise measurement of their weight, e.g., using analytical balances and measuring to 0.0001 g. In small samples the major source of error involved in these two methods, is the blotting method to remove the excess water/liquid. This technique is prone to error however, as it requires a subjective decision by the operator as to how much "excess water is removed". Samples of low apparent porosity (typically < 1 to 2 %) or small size are, therefore more prone to error in their porosity measurements. See e.g, N.A. Pratten, *The precise measurement of the density of small samples,* J. Matls. Sci. **16**, 1737-1747 (1981), for a discussion of errors on measuring density.

Figure 1: Porosity versus density for the Pu-doped samples made from the baseline composition batches Pu68 and Pu67

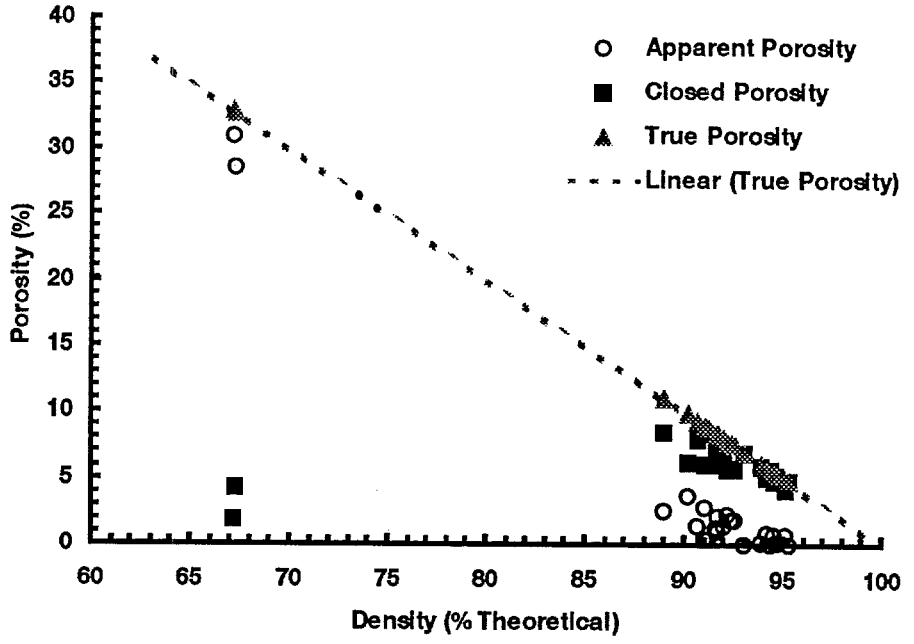


Figure 2: A linear regression fit showing the relationship between apparent porosity and density for the Pu-doped baseline ceramic batches Pu67 and Pu68.

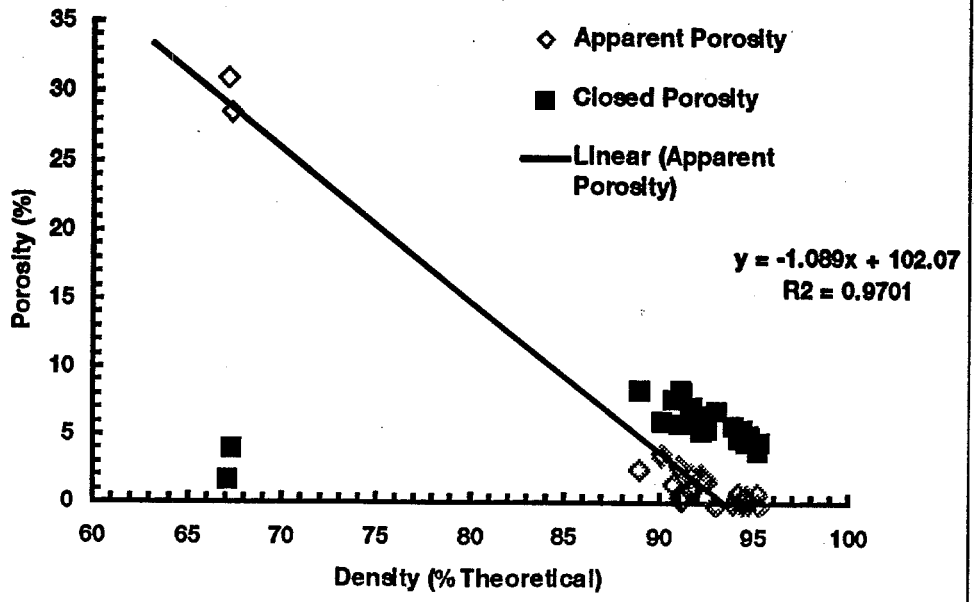


Figure 3: Relationship between open and closed porosity of the Pu-doped baseline composition batches Pu67 and Pu68.

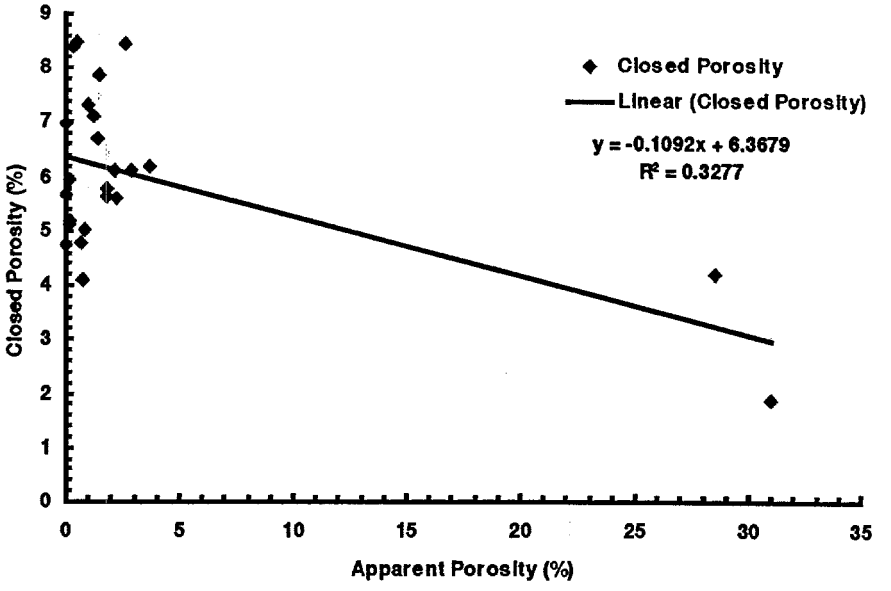


Figure 4: Porosity versus density of the Th-doped Samples made from compositions B1-2, B1-4 and B1-14, Oxide-route wet and dry milled.

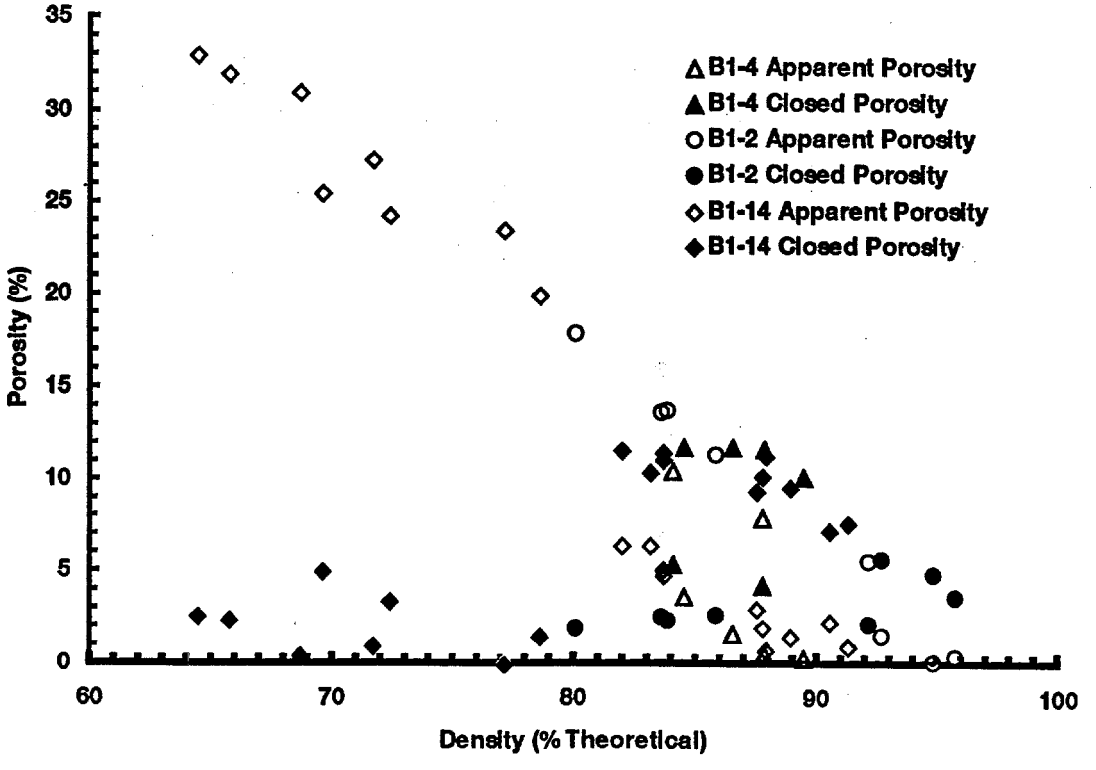
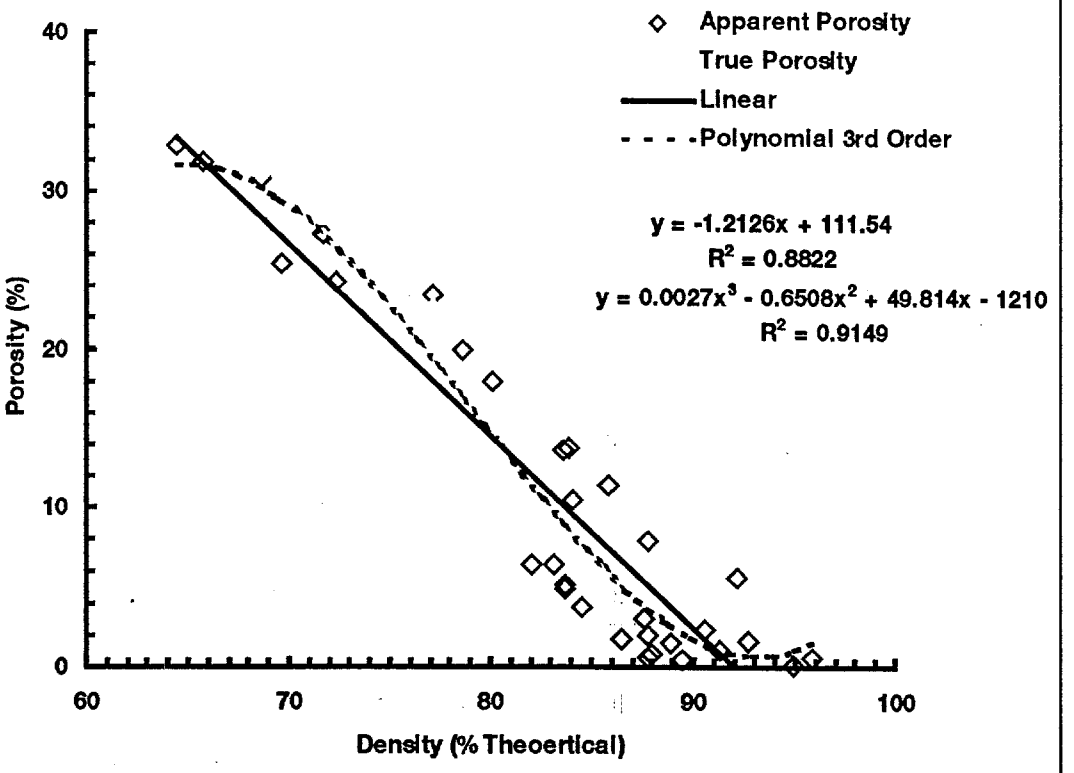


Figure 5: Curve fit of apparent porosity versus density for all the oxide-route batches of compositions B1-2, B1-4 and B1-14



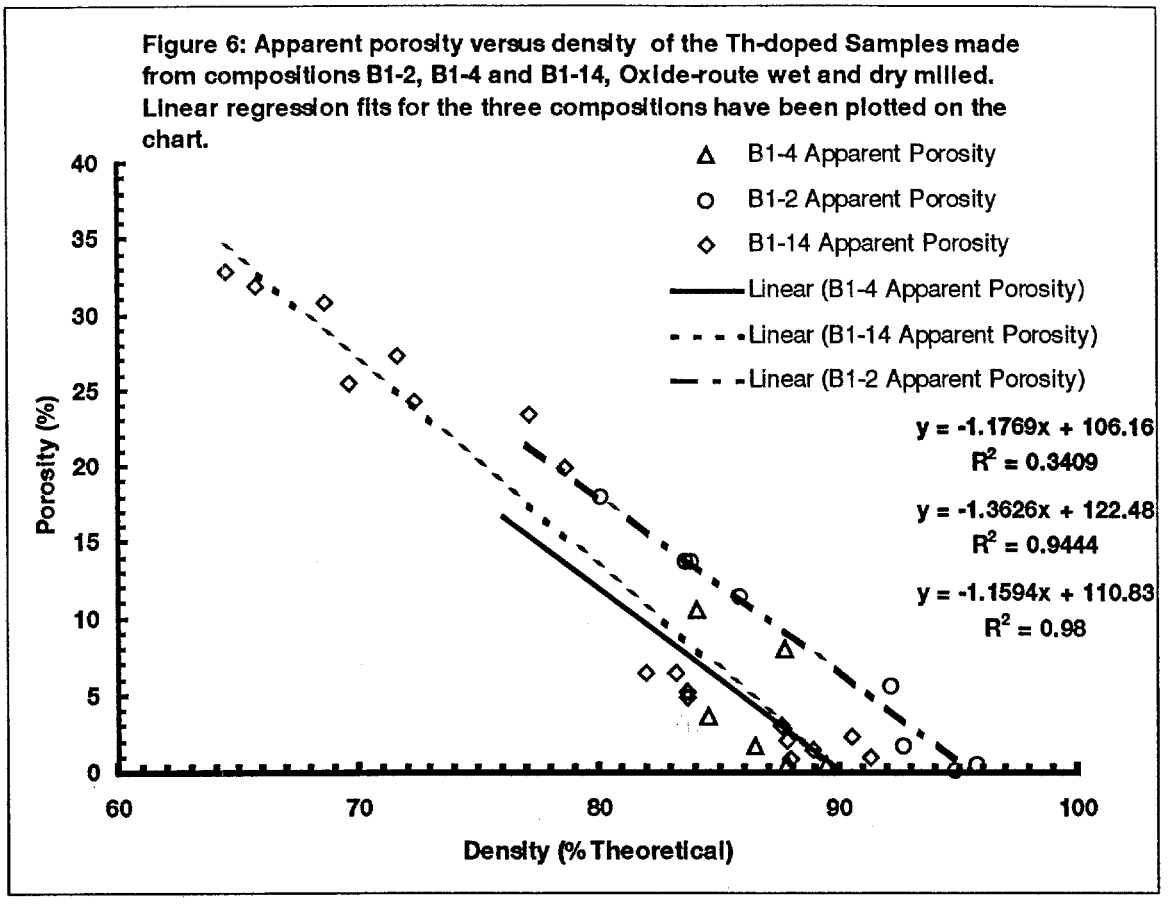
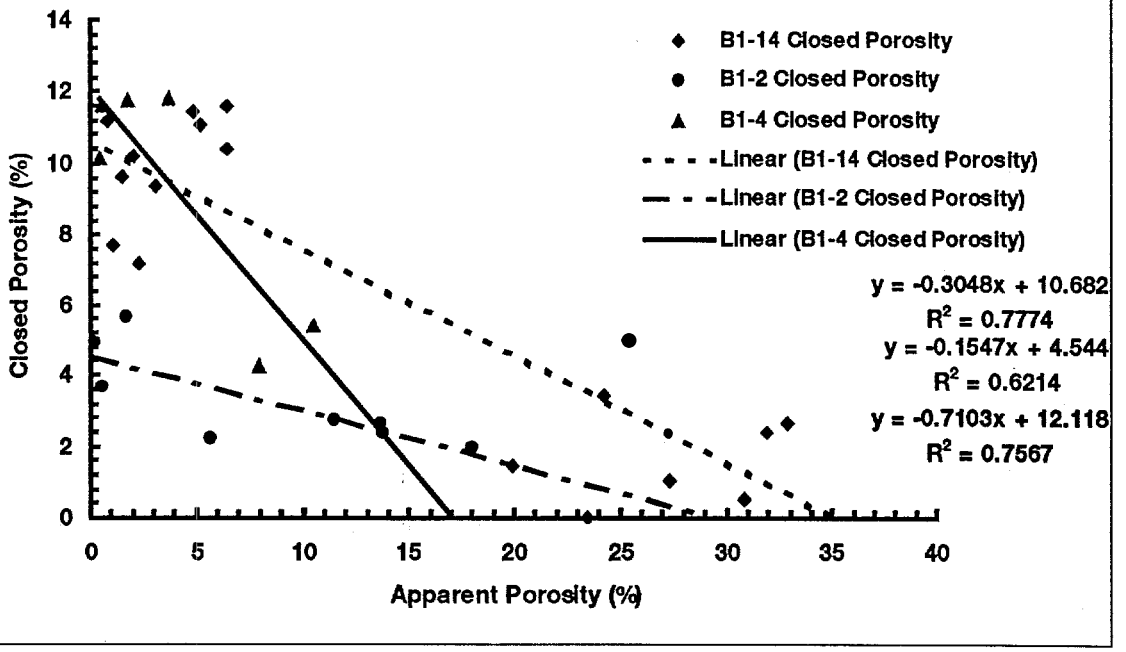


Figure 6: Apparent porosity versus density of the Th-doped Samples made from compositions B1-2, B1-4 and B1-14, Oxide-route wet and dry milled. Linear regression fits for the three compositions have been plotted on the chart.

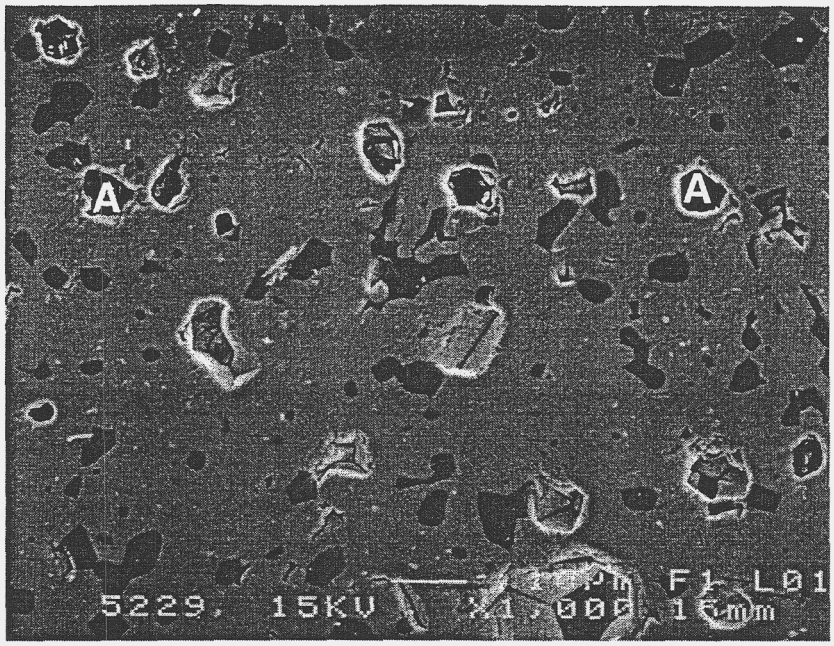
Figure 7: Relationship between apparent and closed porosity for the Th-doped samples



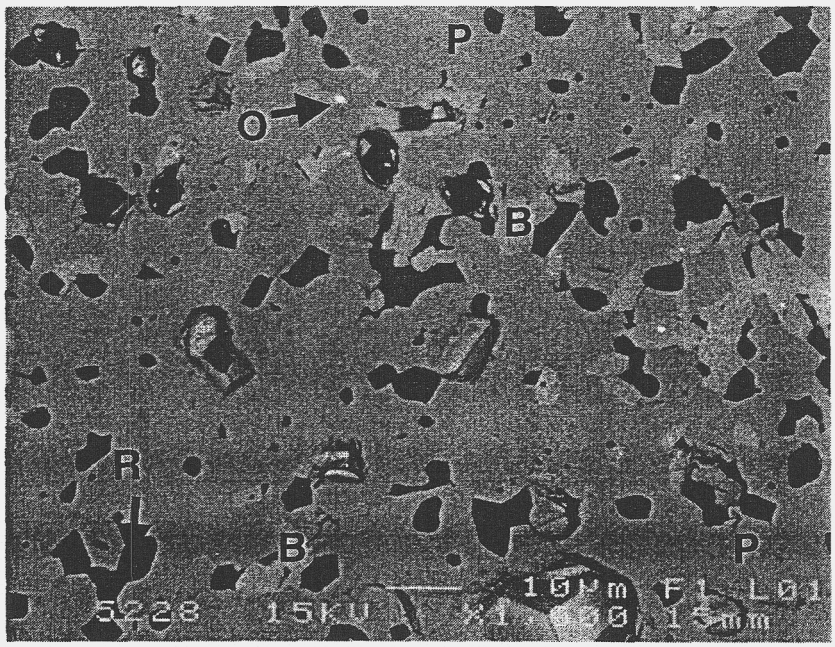
Acknowledgements

Thanks to Dr. A. Day and Mr. S. Leung for their help with the imagie analysis and SEM work.

A APPENDIX A: SEM RESULTS FOR THE PU-DOPED SAMPLES

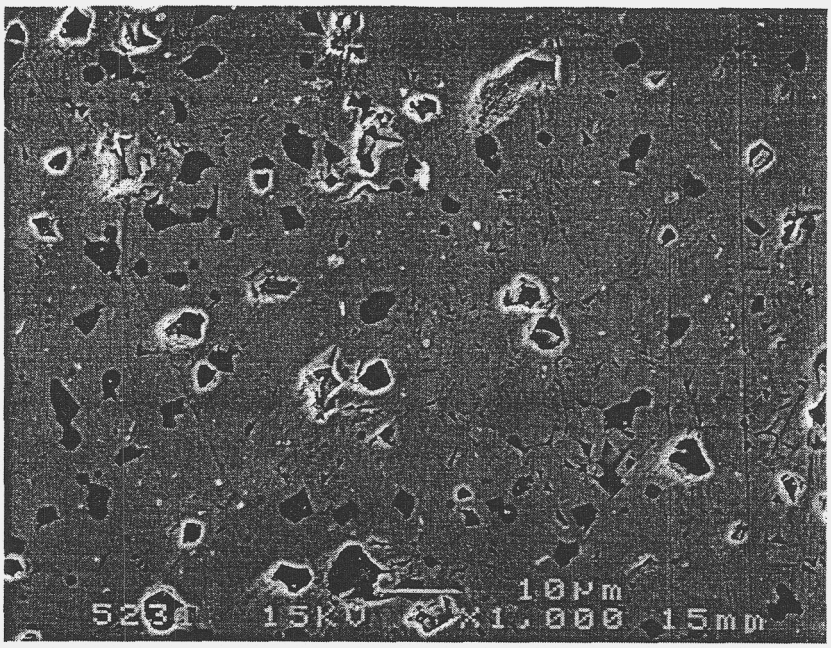


(a)

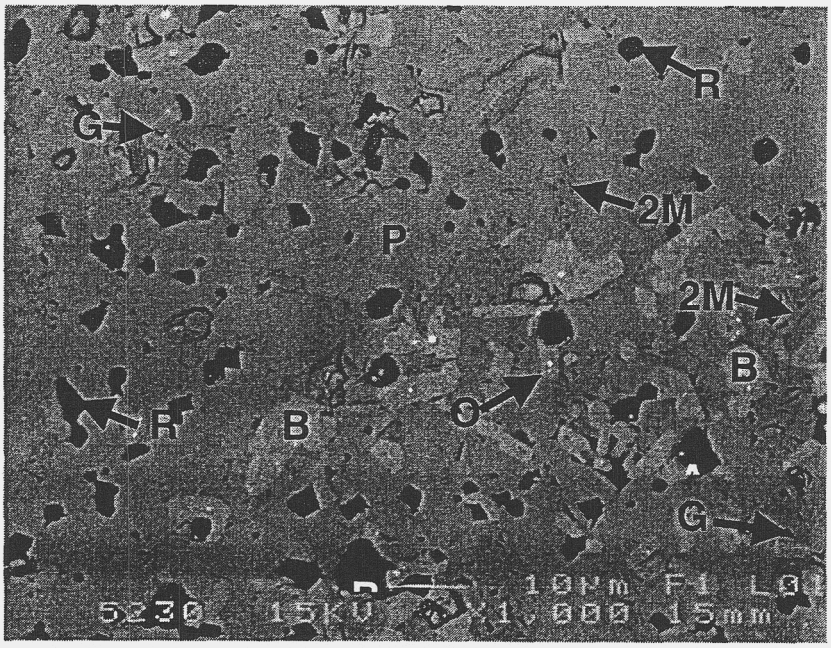


(b) 10 µm.

Figure A-1: (a) Secondary electron micrograph and (b) backscattered electron micrograph of mws980199 (Pu68) (Task 1.2, composition B1-1, oxide-route wet-milled 16 hours, sintered at 1350°C in Ar for 4 hours). The pellet consists of a matrix of pyrochlore (P), with Pu/U-brannerite grains (B, light grey), Hf-doped rutile (R, dark grey), PuO_2 (O, white) and porosity (A).

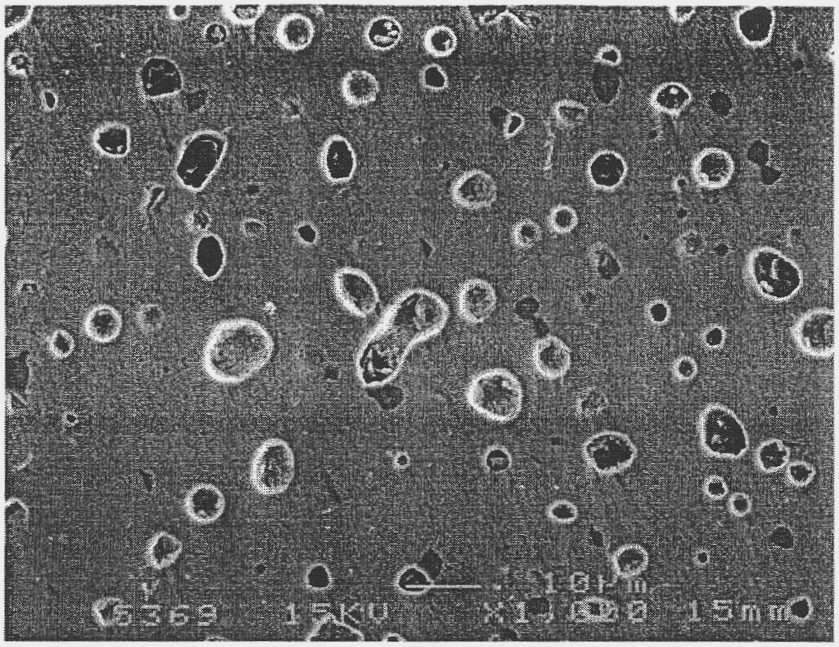


(a)

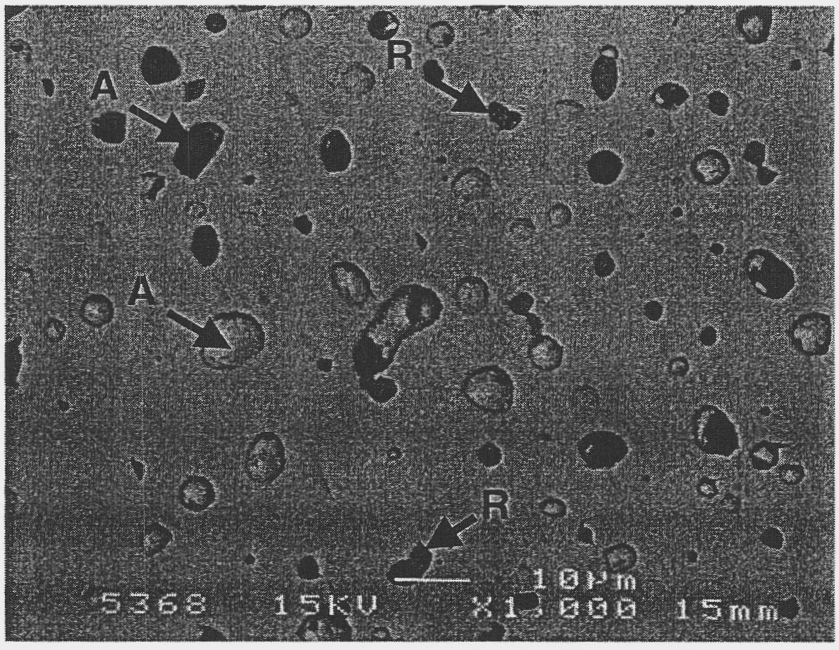


(b) 10 µm.

Figure A-2: (a) Secondary electron micrograph and (b) backscattered electron micrograph of mws980200 (Pu75) (Task 1.2, composition A-7, oxide-route wet-milled, sintered at 1325°C in Ar for 4 hours). The pellet consists of pyrochlore (P), 2M zirconolite (2M, darker grey than pyrochlore), Pu/U-brannerite (B, light grey), Hf-doped rutile (R, dark grey), a silicate intergranular phase (G, black), PuO_2 (O, white spots) and porosity (A).



(a) 10 µm.



(b) 10 µm.

Figure I-12: (a) Secondary electron micrograph and (b) backscattered electron micrograph of mws980361 (Pu105-01A) (Task 1.2, composition B1-13 (~10 % (nominal) perovskite), alkoxide-route, wet-milled 16 hours, sintered at 1350°C in Ar for 4 hours). The matrix is pyrochlore and the dark-grey phase is rutile (R). Porosity (A) is also present. No perovskite or brannerite was detected in this sample.

Table A-1: EDS analyses of phases (number of cations) in the pellets made from the Task 1.2, B1-1, Pu/U-doped wet-milled oxide-route batch. Pellets were sintered in Ar at 1350°C for 4 hours.

Sample No.	Pu68			
	Pyrochlore	Brannerite	Rutile	$(Pu,U)O_2$
Element				
oxygen	7	6	2	2
Ca	0.99	0.08	0.001	0.11
Gd	0.24	0.14	0.002	0.08
Hf	0.22	0.11	0.08	0.03
U	0.41	0.53	0.009	0.42
Pu	0.21	0.21	0.001	0.40
Ti	1.98	2.00	0.91	0.04
Total	4.05	3.08	1.00	1.07

Table A-2: EDS analyses of phases (number of cations) in the pellets made from the Task 1.2, Baseline plus impurities batch Pu75 (composition A-7). The silicate phase was too small to analyse. Pellets were sintered in Ar at 1325°C for 4 hours.

Sample No.	Pu75				
	Pyrochlore	2M Zirconolite	Brannerite	Rutile	$(Pu,U)O_2$
Element					
oxygen	7	7	6	2	2
Ca	0.98	0.76	0.10	0.003	0.13
Gd	0.25	0.17	0.17		0.07
Hf	0.18	0.67	0.09	0.06	0.03
U	0.43	0.16	0.50	0.01	0.43
Pu	0.22	0.08	0.22		0.38
Ti	1.97	1.86	1.99	0.92	0.05
Mg		0.04			
Al	0.03	0.17	0.03	0.006	
Ga		0.03			
K					
Na					
Si					
Ta					
W					
Mo					
Ni					
Fe		0.06			
P					
B					
Cr					
Zn					
Total	4.06	4.01	3.10	1.00	1.08

Note: the absence of a value for an element means that the element is either absent or is present in amounts below the detection limits of the EDS system

Table A-3: EDS analyses of phases (number of cations) in the pellets made from the Task 1.2, B1-13, Pu/U-doped alkoxide-route batch. Pellets were sintered in Ar at 1350°C for 4 hours.

Sample No.	Pu105-01a	
	Pyrochlore	Rutile
Element		
oxygen	7	2
Ca	1.00	0.002
Gd	0.21	
Hf	0.21	0.06
U	0.37	0.01
Pu	0.21	
Ti	2.05	0.93
Total	4.05	1.00

Note: the absence of a value for an element means that the element is either absent or is present in amounts below the detection limits of the EDS system